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FIRST-PRINCIPLES PHASE DIAGRAM OF THE Ce-Th SYSTEM

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1 INTRODUCTION

Actinide physics has seen a remarkable focus the last decade or so due to the combination of improved experimental diamond-anvil-cell techniques and the development of fast computers and more advanced theory. All f -electron systems are expected to have multiphase phase diagrams due to the sensitivity of the f -electron band to external influences such as pressure and temperature. For instance, compression of an f -electron metal generally causes the occupation of f -states to change due to the shift of these bands relative to others. This can in some cases, as in the Ce-Th system, cause the crystal to adopt a lower symmetry structure at elevated pressures. Here we study the phase stabilities of Ce, Th, and the Ce-Th system as a function of compression. Theoretically, both Ce and Th metals are rather well described within the DFT,¹ although a proper treatment of the Ce-Th alloys has not yet been presented.² In the present paper we revisit this problem by applying the modern theory of random alloys based on the coherent potential approximation (CPA).

2 COMPUTATIONAL DETAILS

The calculations we have referred to as exact muffin-tin orbitals (EMTO) are performed using a full-relativistic Green's function technique based on an improved screened KKR method, where the one-electron potential is represented by optimized overlapping muffin-tin (OOMP) potential spheres³. Within the EMTO formalism, the one-electron states are calculated *exactly* for the OOMT potentials. For the exchange/correlation approximation, we use the generalized gradient approximation. For the total energy of random substitutional alloys, the EMTO method has recently been combined with the CPA.³

3 RESULTS

In Figures 1 and 2 we plot the calculated c/a ratio for bct Ce and Th, respectively. Present results agree well with experimental data^{4,5} as well as with those of previous FPLMTO

calculations¹. Figure 3 shows the calculated (EMTO) and measured^{6,7} c/a ratios for the $\text{Ce}_{43}\text{Th}_{57}$ alloy. The results of previous FPLMTO calculations² are also presented.

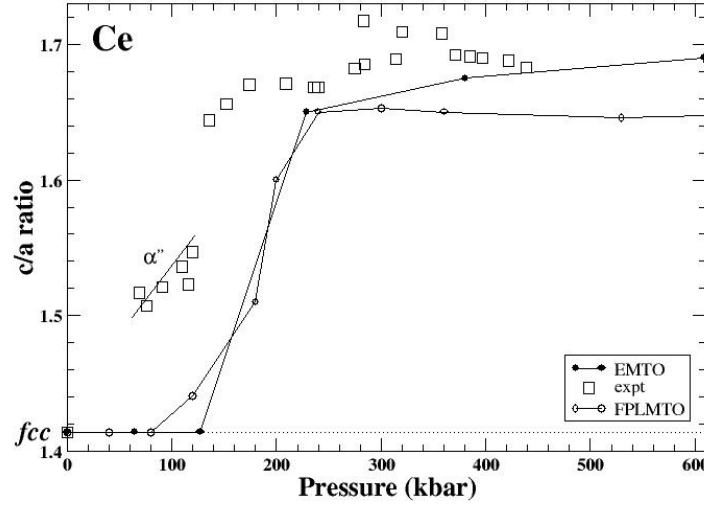


Figure 1 *The c/a axial ratio for the bct structure as a function of pressure for Ce. Experimental data (Ref. 4) are marked with open squares while theoretical results are given by a solid line and filled circles. The results of FPLMTO calculations (Ref. 1) are shown by a solid line and open circles.*

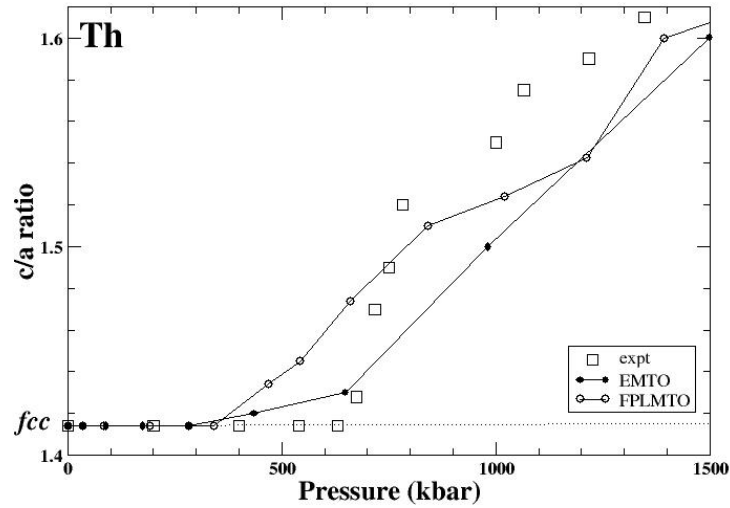


Figure 2 *The c/a axial ratio for the bct structure as a function of pressure for Th. Experimental data (Ref. 5) are marked with open squares while theoretical results are given by a solid line and filled circles. The results of FPLMTO calculations (Ref. 1) are shown by a solid line and open circles.*

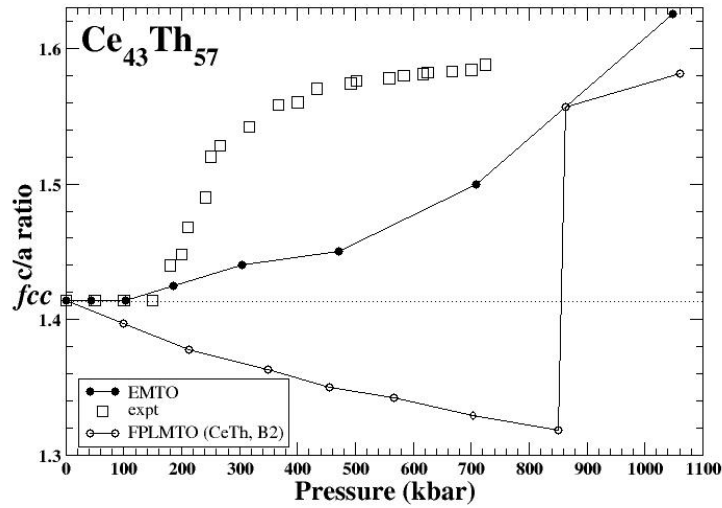


Figure 3 The c/a axial ratio for the bct structure as a function of pressure for $Ce_{43}Th_{57}$ disordered alloy. Experimental data (Ref. 6, 7) are marked with open squares while EMTO theoretical results are given by a solid line and filled circles. Also, the results of FPLMTO calculations (Ref. 2) for Ce-Th-ordered (B2) compound are given by a solid line and open circles.

3 CONCLUSION

We have presented accurate electronic-structure calculations for the Ce-Th system. Generally, the theory reproduced experimental data very well. For the $Ce_{43}Th_{57}$ disordered alloy a CPA treatment is necessary to reproduce the correct structural behaviour.

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